

Quantum-mechanical counterpart of nonlinear optics

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Raman-type laser excitation of a trapped atom allows one to realize the quantum-mechanical counterpart of phenomena of nonlinear optics, such as Kerr-type nonlinearities, parametric amplification, and multimode mixing. Additionally, huge nonlinearities emerge from the interference of the atomic wave function with the laser waves. They lead to a partitioning of the phase space accompanied by a significantly different action of the time evolution in neighboring phase-space zones. For example, a nonlinearly modified coherent “displacement” of the motional quantum state may induce strong amplitude squeezing and quantum interferences. [S1050-2947(97)10406-1]

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I. INTRODUCTION

A single atom trapped in a harmonic potential turns out to be a very well-defined object for studying fundamental phenomena of quantum dynamics. Since the first realization of such a system in an ion trap by Neuhauser *et al.* [1], the subject has stimulated much experimental and theoretical work. As has been shown by Blockley, Walls, and Risken [2], the laser-assisted coupling between the internal and external degrees of freedom of a trapped atom can be described, under appropriate conditions, by a Jaynes-Cummings model. This allows one to study phenomena we are familiar with from cavity QED, such as the micromaser dynamics [3], in the vibronic motion of a trapped atom [4]. Eventually, several proposals have been published for preparing nonclassical states, such as squeezed states [5] and motional number states [6], and successful experiments have been performed [7,8].

The dynamics of a trapped atom, however, not only allows one to reproduce effects of cavity QED in the quantized motion. When the spatial extension of the atomic wave function representing the center-of-mass motion is no longer small compared with the driving laser wavelength, nonlinear effects emerge that have no counterpart in standard nonlinear optics. It has been shown by Vogel and de Matos Filho that the atom may undergo a vibronic coupling, which is very well described by a nonlinear, multiquantum Jaynes-Cummings model [9]. Meanwhile this prediction has been confirmed experimentally [7] and modifications due to micromotion have been studied [10]. The nonlinearities in this model allow us to prepare exciting motional quantum states, such as quantum superpositions of both coherent [11] and squeezed states [12], nonlinear coherent states [13,14], pair coherent states [15] and pair cat states [16]. Measurement techniques for the full diagnostics of motional quantum states have been proposed [17] and realized [18].

These outstanding feasibilities render it possible to raise new types of questions. The nonlinear Jaynes-Cummings model has introduced new kinds of nonlinearities that substantially modify phenomena we are familiar with from nonlinear optics, such as multiphoton absorption and emission.

In nonlinear optics, however, other interactions are known which leave the electronic transitions of the nonlinear medium almost unchanged. Examples are the Kerr nonlinearity, parametric interactions, and several types of nonlinear wave mixings. The question appears as to whether it is possible to realize such phenomena in the motional dynamics of a single atom, where the trap potential replaces a cavity used in nonlinear optics.

In the present contribution we propose Raman-type excitations for inducing various kinds of nonlinear interactions in the quantized motion of a trapped atom. We consider the quantum-mechanical counterpart of nonlinear optical effects that do not influence the electronic degrees of freedom of the atomic medium. We show that even a single degree of freedom of the atomic center-of-mass motion can be driven in a strongly nonlinear manner. Surprising phenomena are caused by the interference effects of the atomic wave function with the driving light waves. They induce a nonlinear partitioning of the phase space, the action of the time evolution being different in neighboring phase-space zones. This partitioning may be used for the generation of nonclassical effects, such as amplitude squeezing and quantum interferences.

The paper is organized as follows. In Sec. II the basic model for the Raman-induced motional dynamics is introduced and the effective Hamiltonian for the nonlinear motional interactions is derived. Section III is devoted to the nonlinear phase-space partitioning together with the illustration of its effects in simple examples of motional dynamics. A summary and some conclusions are given in Sec. IV.

II. RAMAN-INDUCED MOTIONAL DYNAMICS

Let us consider an atom harmonically bound in a trap. In general, the atom oscillates in the three principal axes of the trap with frequencies ν_i ($i=1,2,3$). The trapped atom is driven in a Raman configuration with two classical laser fields of frequencies ω_L and $\omega_L + \Delta$ ($\Delta \ll \omega_L$), which are off-resonant with respect to the electronic transitions, see Fig. 1. During the interaction with the two lasers, the atom stays in its electronic ground state. However, in the resolved sideband regime and for appropriately chosen laser-beam ge-

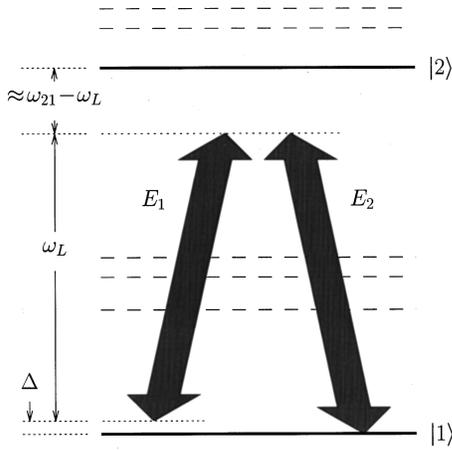


FIG. 1. The $|1\rangle \leftrightarrow |2\rangle$ transition of a trapped atom is driven by two off-resonant laser fields E_1 and E_2 of frequencies ω_L and $\omega_L + \Delta$, respectively. Other electronic states (broken lines) are far off-resonant. The beat frequency Δ can be tuned on resonance with multiples of vibrational frequencies.

ometry and laser detuning Δ , it is possible to affect the motional quantum state of the atom in a well-controlled manner.

The effective interaction Hamiltonian for the Raman coupling (in optical rotating-wave approximation) reads as

$$\hat{H}_L(t) = \frac{1}{2} \hbar \Omega e^{-i[\Delta t - \mathbf{k} \cdot \hat{\mathbf{r}}]} + \text{H.c.}, \quad (1)$$

where $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$ is the difference wave vector of the two laser beams and $\hat{\mathbf{r}}$ is the operator of the atomic center-of-mass position. For small relative detunings from the frequency ω_{21} of the dipole transition ($|\omega_{21} - \omega_L|/\omega_{21} \ll 1$), the effective two-photon Rabi frequency Ω is given by

$$\Omega = \frac{1}{2} \frac{\Omega_1 \Omega_2^*}{\omega_{21} - \omega_L}, \quad (2)$$

with $\Omega_i = 2dE_i/\hbar$ ($i = 1, 2$) being the single-photon Rabi frequencies of the dipole transition of dipole moment d , driven by the electric-field amplitudes E_1 and E_2 of the two lasers. The phase of $\Omega = |\Omega|e^{i\varphi}$ is determined by the difference phase of the two laser fields $\varphi = \varphi_1 - \varphi_2$ and can be held very stable in experiments. Equation (1) can be written in terms of creation and annihilation operators of vibrational quanta by using the relations $k_i \hat{x}_i = \eta_i(\hat{a}_i + \hat{a}_i^\dagger)$, where k_i are the projections of the wave-vector difference on the principal axes x_i of the trap and η_i are the Lamb-Dicke parameters of the vibration in these directions. After disentangling the resulting exponential operator function, the Hamiltonian (1) may be expanded in a power series as

$$\begin{aligned} \hat{H}_L(t) &= \frac{1}{2} \hbar \Omega e^{-i\Delta t} e^{-(\eta_1^2 + \eta_2^2 + \eta_3^2)/2} \\ &\times \sum_{m,m'} \sum_{n,n'} \sum_{l,l'} \frac{(i\eta_1)^{m+m'} (i\eta_2)^{n+n'} (i\eta_3)^{l+l'}}{m!m'!n!n'!l!l'!} \\ &\times \hat{a}_1^{\dagger m} \hat{a}_2^{\dagger n} \hat{a}_3^{\dagger l} \hat{a}_1^m \hat{a}_2^{n'} \hat{a}_3^{l'} + \text{H.c.} \end{aligned} \quad (3)$$

This interaction includes, via the mode functions [cf. Eq. (1)] of the laser waves, a laser-assisted coupling of the three motional degrees of freedom (x_1, x_2, x_3). Since the wave-vector

difference \mathbf{k} is determined by the laser-beam geometry, the coupling of the motional degrees of freedom can be designed to include one, two, or three directions.

To consider these couplings in more detail, we assume that the vibrational frequencies are well resolved by the Raman excitation, so that we may introduce a vibrational rotating-wave approximation. Choosing the laser beat frequency to be a multiple of the three vibrational frequencies, $\Delta = s_1 \nu_1 + s_2 \nu_2$ ($s_{1,2} = 0, \pm 1, \pm 2, \dots$), one obtains a coupling of all vibrational modes [19]. In this case the interaction Hamiltonian (in the interaction picture) is of the form [20]

$$\begin{aligned} \hat{H}_{\text{int}} &= \frac{1}{2} \hbar \Omega \sum_{n=-\infty}^{\infty} \hat{g}_{n-s_1}(\hat{a}_1^\dagger, \hat{a}_1; \eta_1) \hat{g}_{n-s_2}(\hat{a}_2^\dagger, \hat{a}_2; \eta_2) \\ &\times \hat{g}_n(\hat{a}_3^\dagger, \hat{a}_3; \eta_3) + \text{H.c.} \end{aligned} \quad (4)$$

and the operator-valued functions $\hat{g}_k(\hat{a}^\dagger, \hat{a}; \eta)$ are given by

$$\hat{g}_k(\hat{a}^\dagger, \hat{a}; \eta) = \begin{cases} (i\eta \hat{a}^\dagger)^{|k|} \hat{f}_{|k|}(\hat{n}; \eta) & \text{if } k \geq 0 \\ \hat{f}_{|k|}(\hat{n}; \eta) (i\eta \hat{a})^{|k|} & \text{if } k < 0. \end{cases} \quad (5)$$

The Hermitian operator functions $\hat{f}_k(\hat{n}; \eta)$ depend solely on the number of vibrational quanta $\hat{n} = \hat{a}^\dagger \hat{a}$ and read (in normally ordered form) as

$$\hat{f}_k(\hat{n}; \eta) = e^{-\eta^2/2} \sum_{l=0}^{\infty} \frac{(-1)^l \eta^{2l}}{l!(l+k)!} \hat{a}^{\dagger l} \hat{a}^l. \quad (6)$$

From Eqs. (5) and (6) it is seen, that for the decreasing Lamb-Dicke parameter only the coupling with $k=0$ survives. Therefore, by varying the geometry of the laser-beam propagation one can vary the Lamb-Dicke parameters in order to change the Hamiltonian from a coupling of only one, two, or three vibrational modes.

It is seen from Eqs. (4) and (5) that the Hamiltonian describes a motional dynamics with the following basic effects. First, there appear combinations of different powers of the motional operators $\hat{a}_i, \hat{a}_i^\dagger$. Interactions of this type represent the quantum-mechanical counterpart of wave-mixing effects in nonlinear optics. Second, via the functions $\hat{f}_k(\hat{n}; \eta)$ the couplings depend in a nonlinear manner on the excitations of the modes. This results from the interference of the atomic (center-of-mass) wave functions and the beat node of the laser waves, which is a typical effect of quantized atomic motion.

III. NONLINEAR PHASE-SPACE PARTITIONING

To get some insight into these effects, we first consider the one-dimensional dynamics, where only the motion in x_1 direction is affected by the lasers ($\eta_2 = \eta_3 = 0$). This requires a geometry of laser propagations with vanishing projections of the difference wave-vector \mathbf{k} on the axes x_2 and x_3 . In this case the Hamiltonian simplifies as

$$\hat{H}_{\text{int}} = \frac{1}{2} \hbar \Omega \hat{f}_k(\hat{n}; \eta) (i\eta \hat{a})^k + \text{H.c.}, \quad (7)$$

where we assumed a laser detuning of $\Delta = k\nu_1$ ($k \geq 0$) and we have omitted the indices of the x_1 direction. Interactions

of this type may be considered as nonlinear mode couplings of one (weakly excited) quantized mode with (strongly excited) classical modes. Such approximations are frequently used in quantum optics. Experiments of the type proposed here would allow one to realize these couplings almost perfectly and to study the additional (excitation-dependent) nonlinearities.

For example, let us consider the one-quantum resonance ($\Delta = \nu_1$) in more detail. In this case the structure of the unitary time-evolution operator obtained from the Hamiltonian (7) shows some formal resemblance to a nonlinearly modified coherent “displacement” operator [21],

$$\begin{aligned} \hat{U}_{\text{int}}(t) &= \hat{D} \left[-\frac{\eta\Omega^*t}{2} \hat{f}_1(\hat{n}; \eta) \right] \\ &= \exp \left[-\frac{\eta\Omega^*t}{2} \hat{a}^\dagger \hat{f}_1(\hat{n}; \eta) + \frac{\eta\Omega t}{2} \hat{f}_1^\dagger(\hat{n}; \eta) \hat{a} \right]. \end{aligned} \quad (8)$$

For small values of the Lamb-Dicke parameter, $\eta \ll 1$, according to Eq. (6) the operator (8) may be replaced by the usual displacement operator $\hat{D}(-\eta\Omega^*t/2)$.

The nonlinear dependence of the “displacement” operator (8) on the mean number of vibrational quanta leads to effects of a new type. For a first insight we may replace the number operator by its eigenvalue. We arrive at the c -number function $f_1(n; \eta) = \langle n | \hat{f}_1(\hat{n}; \eta) | n \rangle$, which reads as

$$f_1(n; \eta) = \frac{e^{-\eta^2/2}}{n+1} L_n^{(1)}(\eta^2), \quad (9)$$

with $L_n^{(k)}(x)$ being Laguerre polynomials. To consider the action of the nonlinear displacement in phase space, it is advantageous to introduce the (complex) phase-space amplitude α by setting $n = |\alpha|^2$. The resulting function $f_1(|\alpha|^2; \eta)$ has zeros and changes its sign for certain values of $|\alpha|$. Consequently, the direction of the displacement can be reversed, depending on the amplitude of the quantum state in phase space. That is, the phase space is effectively partitioned in zones. The action of the displacement in adjacent zones differs in the fact that the directions of displacements are opposite to each other, along an axis which is controlled by the phase difference of the lasers. These phase-space zones are separated by the circles on which the coupling function $f_1(|\alpha|^2; \eta)$ changes its sign. This nonlinear partitioning of the phase space leads to striking consequences with respect to the evolution of the quantum state.

Let us consider the evolution of a coherent state that is initially located on the boundary between two such phase-space zones. Inside the corresponding circle the coupling $f_1(|\alpha|^2; \eta)$ is positive and outside it is negative. Due to this fact the nonlinear “displacement” operator tends to split the coherent state as shown in Fig. 2. For rather short times the state can exhibit a significant reduction of phase fluctuations. In the further course of time the states are split into well separated substates. This leads to a coherent superposition of two quantum states, accompanied by quantum-interference effects. The displacement of each substate is limited by the boundaries between the phase-space zones, where the

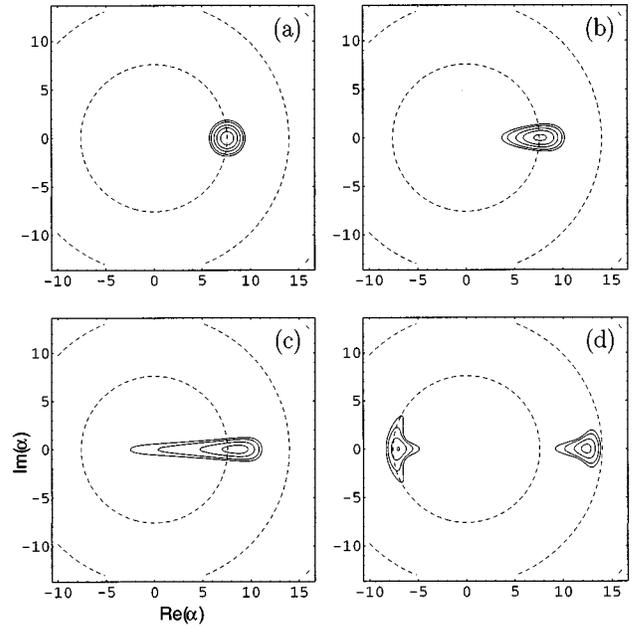


FIG. 2. Time evolution of a coherent state that is initially placed on the boundary between two phase-space zones with opposite displacement directions (chosen along the real axis). The dimensionless times $\eta|\Omega|t$ are given by 0 (a), 2.5 (b), 5 (c), and 15 (d); $\eta = 0.25$. The contours represent the Q functions of the motional quantum states.

strength of displacement becomes negligible. The result is a squeezing of each substate onto the corresponding circle partitioning the phase space.

This effect can be used to generate quantum states exhibiting strong amplitude squeezing. Let us consider the nonlinear displacement of a coherent state that is initially located within a single phase-space zone. As expected, the state is displaced in a well-defined direction in phase space until it is squeezed onto the next circle separating two zones. The result consists in a strongly amplitude-squeezed state [22] with a nonvanishing coherent amplitude as shown in Fig. 3. It is worth noting that in its further evolution this quantum state does not approach a Fock state. The reason consists in the

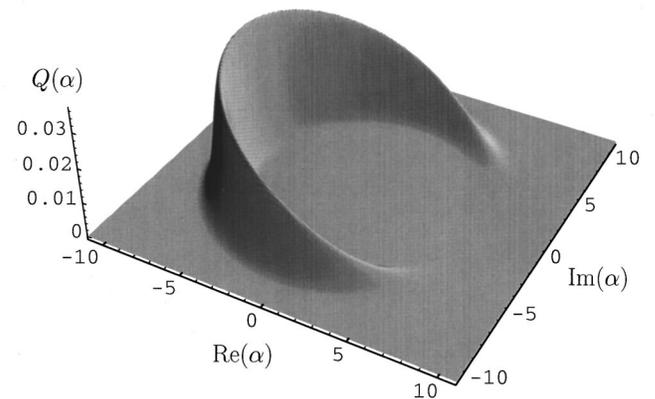


FIG. 3. Q function of a strongly amplitude-squeezed state with $\langle \Delta \hat{n}^2 \rangle / \langle \hat{n} \rangle = 0.006$. This state is reached from an initially coherent state ($\alpha = -9$) in a dimensionless time $\eta|\Omega|t \approx 10$, for $\eta = 0.25$. The displacement acts along the real axis.

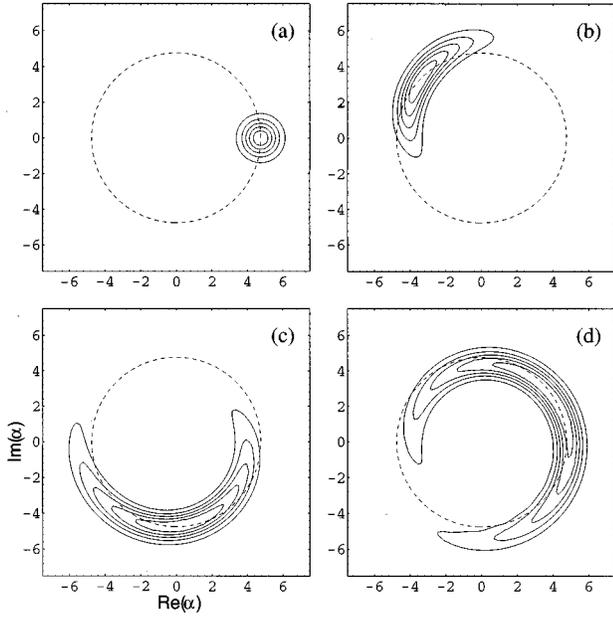


FIG. 4. Time evolution of the Q function for $k=0$ (Kerr-type effects) and $\eta=0.25$. The dimensionless times $|\Omega|t$ are (a) 0, (b) 173.5, (c) 346.6, and (d) 500.

fact that, in general, the transitions between neighboring phase-space zones are very weak, but not suppressed completely. This leads to continued deformations of the phase-space distributions of the motional quantum state.

The one-dimensional Hamiltonian (7) allows us to consider other types of phenomena known from nonlinear optics. Choosing $k=0$, the corresponding dynamics is related to the Kerr nonlinearity [23]. The standard Kerr nonlinearity is reproduced by expanding the Hamiltonian up to η^4 . In the more general case of larger Lamb-Dicke parameters the nonlinear function $f_0(n; \eta) = \langle n | \hat{f}_0(\hat{n}; \eta) | n \rangle$ plays a similar role as the function $f_1(n; \eta)$ for the case $k=1$. Its oscillations as a function of n again lead to the phase-space partitioning effect. This is illustrated in Fig. 4 for an initially coherent state situated at a circle in phase space where $f_0(n; \eta) = 0$. One clearly observes a rotation of the state which is due to the term $\propto \eta^2$ of $\hat{f}_0(\hat{n}; \eta)$. Moreover, the state is significantly deformed: inside and outside the circle the state undergoes phase shifts into opposite directions, reflecting the change in sign of the coupling.

For $k=2$ the Hamiltonian (7) represents the nonlinear generalization of a classically driven parametric interaction. For $\eta \ll 1$ the time-evolution operator agrees with the squeeze operator. This limiting case has been realized experimentally [7]. In the more general case of larger Lamb-Dicke parameters, a rather complex dynamics appears. The interpretation of all of its features needs some further research.

For studying a quantized version of the parametric interaction, the coupling of two degrees of freedom is needed. Consider a laser-beam geometry with the projection of the difference wave-vector \mathbf{k} on the x_3 axis being zero, so that $\eta_3=0$. The dynamics couples the motion in x_1 and x_2 direc-

tions. For example, a detuning of $\Delta = 2\nu_1 - \nu_2$ ($s_1=2, s_2=-1$) reduces the interaction Hamiltonian (4) to

$$\hat{H}_{\text{int}} = -\frac{i}{2} \hbar \eta_1^2 \eta_2 \Omega \hat{f}_2(\hat{n}_1; \eta_1) \hat{a}_1^2 \hat{a}_2^\dagger \hat{f}_1(\hat{n}_2; \eta_2) + \text{H.c.}, \quad (10)$$

representing a nonlinear generalization of the parametric interaction. For small Lamb-Dicke parameters, $\eta_{1,2} \ll 1$, this interaction simplifies to

$$\hat{H}_{\text{int}} = -\frac{i}{2} \hbar \eta_1^2 \eta_2 \Omega \hat{a}_1^2 \hat{a}_2^\dagger + \text{H.c.}, \quad (11)$$

which is the standard form of the parametric coupling. Beyond the Lamb-Dicke regime the interaction includes nonlinearities of the type considered above, which now appear in both motional degrees of freedom. Consequently, the nonlinear phase-space partitioning effects considered above will be of relevance for each degree of freedom involved in the Raman-induced motional dynamics.

IV. SUMMARY AND CONCLUSIONS

In conclusion, we have shown that a Raman-type laser excitation allows one to induce nonlinear interactions of motional degrees of freedom of a trapped atom, which are closely related to phenomena of nonlinear optics that do not change the electronic quantum states of the medium. The number of coupled modes can be easily controlled by the laser-beam geometry. Standard effects can be realized, including coherent displacements, Kerr nonlinearities, and parametric mode couplings. In the laser-assisted motional dynamics additional nonlinearities emerge, which are caused by the interference between the light waves and the wave function representing the atomic center-of-mass motion.

An important consequence of these nonlinearities consists in a partitioning of the motional phase space, which is caused by an oscillatory behavior of the motional interactions as a function of the phase-space amplitude. In neighboring phase-space zones the actions of the time evolution appear to be significantly different from each other. For example, in two adjacent zones a nonlinearly modified ‘‘displacement’’ operator acts in opposite directions. Consequently, a quantum state whose initial location is on the boundary between two zones will be split in two substates, which eventually gives rise to quantum interferences. Moreover, the partitioning allows one to generate strongly amplitude-squeezed motional states. Eventually, in the case of a generalized Kerr nonlinearity the phase-space partitioning may lead to pronounced deformations of the initial state, which are caused by opposite phase shifts appearing in adjacent phase-space zones.

The phase-space partitioning, although illustrated in this paper for the motional dynamics in one dimension, is a universal feature of the interference between the Raman beat node and the wave function describing the center-of-mass motion of the atom. When two or three dimensions are involved in the Raman-induced dynamics, the partitioning effects appear in the phase space of each motional degree of freedom. Consequently, the coupling between different motional modes will be strongly influenced by the interplay of these nonlinear effects. In general the dynamics will sensi-

tively depend on the initial conditions. Besides the feasibility of realizing phenomena well known from nonlinear optics in the motion of a trapped atom, this opens novel possibilities for studying nonlinear phenomena in a well-defined quantum system.

Note added in proof: Recently we became aware of the fact that G. S. Agarwal and J. Banerji [Phys. Rev. A (this issue) **55**, 4007 (1997)] have proposed a Raman excitation

scheme for inducing a parameteric interaction of motional degrees of freedom of a trapped ion.

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- [19] Since $\nu_3 \approx \nu_1 + \nu_2$ holds for a quadrupole trapping potential, this choice describes all possible combinations of the three vibrational frequencies.
- [20] We consider here the nondegenerate case $\nu_1 \neq \nu_2$. For the degenerate case only one vibrational frequency is needed to completely describe a resonant detuning: $\Delta = s\nu_1$. In this case the Hamiltonian is given by Eq. (4) with an additional sum ($\sum_{s_1=-\infty}^{\infty}$) over s_1 while replacing $s_2 \rightarrow s - s_1$. This sum reflects the manifold of possible resonances, generated by the degeneracy $\nu_1 = \nu_2$.
- [21] We would like to point out that the nonlinear "displacement" operator defined in Eq. (8) does not exhibit the standard properties of a displacement operator, which is a consequence of the fact that the operator-valued function \hat{f}_1 does not commute with the annihilation and creation operators. However, its action shows some resemblance to a nonlinear displacement that depends on the amplitude of the quantum state in phase space.
- [22] Note that a hint on such an amplitude-squeezing effect has already been found experimentally; see footnote 36 of Ref. [8].
- [23] A different scheme to realize Kerr-type effects of atomic motion has been proposed by J. K. Breslin, C. A. Holmes, and G. J. Milburn (unpublished).